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**Japanese Patent Application JP 11 – 343353 A**

**Polyester Resin Sheeting and Manufacturing Method**

**Your Ref: No. 1256**

**For: Eastman Chemical Company**

9109 Eagle Hills Dr . Las Vegas NV 89134 . ph: 866-722-7990 (toll-free) . fax: 702-973-6009 . email: sales@tls-translations.com

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	(72) Inventor: Sugano HIROSHI
	Sumitomo Bakelite Co., Ltd.
	5-8 Higashi-shinagawa 2-chome
	Shinagawa-ku, Tokyo

**(54) [Title of the Invention]**

**Polyester Resin Sheeting and Manufacturing Method**

**(57) [Summary]**

**[Problem]**

To provide polyester resin sheeting that can be calendar-molded without adhering to the calendering rolls.

**[Means of Achievement]**

A polyester resin sheeting that contains 0.1 to 2 weight parts of a fatty acid ester lubricant per 100 weight parts of a copolyester resin in which the ethylene glycol component of a polyethylene terephthalate resin is substituted with 10 to 40 mol% cyclohexanedimethanol.

## **[Claims]**

**[Claim 1]** Polyester resin sheeting, characterized in containing 0.1 to 2 weight parts of a fatty acid ester lubricant per 100 weight parts of a copolyester resin in which the ethylene glycol component of a polyethylene terephthalate resin is substituted with 10 to 40 mol% cyclohexanedimethanol.

**[Claim 2]** Polyester resin sheeting, characterized in containing 1 to 20 weight parts of a white pigment and 0.1 to 2 weight parts of a fatty acid ester lubricant per 100 weight parts of a copolyester resin in which the ethylene glycol component of a polyethylene terephthalate resin is substituted with 10 to 40 mol% cyclohexanedimethanol.

**[Claim 3]** A method for manufacturing polyester resin sheeting, characterized in that the polyester resin composition according to claim 1 or 2 is calendered.

**[Claim 4]** Sheetting for cards for which is used the polyester resin sheeting according to Claim 2.

**[Claim 5]** Sheetting for cards, characterized in that at least one side of the polyester resin sheeting according to Claim 1 or 2 is embossed to a surface roughness Ra of 0.5 to 15  $\mu\text{m}$ .

## **[Detailed Description of the Invention]**

### **[0001]**

**[Technological Field of the Invention]** The present invention relates to polyester resin sheeting molded by means of calendaring, and in particular relates to polyester resin sheeting having a specific composition that is able to be molded into sheeting in a high-productivity calendaring process without adhering to the calendaring rolls.

### **[0002]**

**[Prior Art]** Applications involving polyester resin sheetings have expanded in recent years to include casings and containers for electronic, electrical, and office equipment; packaging materials for food products, pharmaceuticals, and the like; lids for molded containers, containers for blister packaging, and other materials; sheeting for construction materials; IC cards manufactured by means of the layering of core sheeting and overlay sheeting and equipped with modules having internally mounted IC chips and the like; and magnetic strip cards, ID cards, and other card sheeting. These polyester resin sheetings have conventionally been molded into sheetings as a result of molten resin pellet extrusion. However, such extrusion molding methods

produce sheeting at low speeds, have inadequate productivity, and produce insufficient thickness precision.

[0003] On the other hand, vinyl chloride resin sheeting has conventionally been molded by means of calendering and extrusion molding. Advantages of calender molding include a high-speed sheeting production and excellent thickness precision. With sheeting is to be calendered from polyester resins, the sheeting tightly adheres to the calendering rolls due to the properties of the polyester resin, making the molding of such sheeting effectively impossible. If the temperature of the calendering rolls is reduced, the surface layer of the resulting sheeting will be roughened, which also makes the molding of such sheeting effectively impossible.

[0004]

**[Problems to Be Solved by the Invention]** It is an object of the present invention to provide polyester resin sheeting that can be molded by means of high-productivity calendering without adhering to the calendering rolls.

[0005]

**[Means of Achievement]** As a result of investigations aimed at attaining the object stated above, it was discovered that the adhering of a polyester resin to a calendering roll can be prevented if a certain degree of lubricity is provided when the polyester resin and the calendering rolls are in contact with each other. Based on this knowledge, further studies were conducted, and it was concluded that (1) it is difficult to impart lubricity to the calendering rolls themselves; (2) it is consequently necessary to impart lubricity to the sheeting to be molded; and (3) in order to impart lubricity to the sheeting itself, a lubricity-imparting agent can be added in advance to the polyester resin. Polyethylene terephthalate resin is the most common polyester resin, but it is difficult to calender because of its crystallinity. As a result of various investigations, it was learned that only a copolyester resin in which the ethylene glycol component of a polyethylene terephthalate resin had been substituted with 10 to 40 mol% cyclohexanedimethanol could become amorphous. Furthermore, as a result of diligent investigations into lubricity-imparting agents that could be added to polyester resins, it was learned that fatty acid ester lubricants have good compatibility with polyester resins, are readily miscible with these resins, do not degrade the sheeting molding properties of polyester resins, provide the resulting sheeting with good

characteristics (appearance, physical properties, etc.), and have excellent effects in terms of calendering properties.

**[0006]** The present invention is based on the discoveries described above, and provides polyester resin sheeting that contains 0.1 to 2 weight parts of an fatty acid ester lubricant per 100 weight parts of a copolyester resin in which the ethylene glycol component of a polyethylene terephthalate resin is substituted with 10 to 40 mol% cyclohexanedimethanol, and also provides polyester resin sheeting obtained as a result of adding a white pigment to the aforementioned polyester resin composition in an amount of 1 to 20 weight parts per 100 weight parts of the copolyester resin. The invention further constitutes a method for manufacturing polyester resin sheeting by means of calendering the aforementioned polyester resin composition, and sheeting for cards in which at least one side of the polyester resin sheeting is embossed to a surface roughness Ra of 0.5 to 15  $\mu\text{m}$ .

**[0007]**

**[Embodiment of the Invention]** The present invention is explained in detail below. A copolyester resin in which the ethylene glycol component of a polyethylene terephthalate resin is substituted with 10 to 40 mol% cyclohexanedimethanol is used as the polyester resin in the present invention. If less than 10 mol% of the ethylene glycol component is substituted, the temperature limits that can be set during calendering will be narrow due to crystallinity, and the material will become milky and unusable. Conversely, if more than 40 mol% of the ethylene glycol component is substituted, calendering will become difficult due to crystallinity, the tensile strength and the bending elastic modulus will decrease, and the sheeting will become less elastic and unusable. Therefore, the replacement amount of ethylene glycol is 10 to 40 mol%, and preferably 20 to 35 mol%.

**[0008]** Examples of compounds that may be used for the fatty acid ester lubricant to be blended with the above-mentioned polyester resin include butyl stearate, cetyl palmitate, stearic acid monoglyceride, stearic acid diglyceride, stearic acid triglyceride, montan wax acid esters, montanic acid esters partially saponified with calcium, low esters, and aliphatic dicarboxylic acid esters. These compounds can be used alone or in mixtures of two or more thereof.

**[0009]** If the blending ratio of the fatty acid ester lubricant is too low, no blending effect will be apparent, and the polyester resin will unavoidably adhere to the calendering rolls. Conversely, if

the ratio is too high, sheeting transparency will be compromised, and the aliphatic acid lubricant will often migrate to the sheeting surface (blooming), which is undesirable in the end product. Consequently, the aliphatic acid lubricant is used in the present invention in the amount of 0.1 to 2 weight parts, and preferably 0.3 to 1 weight parts, per 100 weight parts of the above-mentioned copolyester resin.

**[0010]** According to the present invention, it is also possible to use a variety of commonly blended additives other than the fatty acid ester lubricants. Examples include other lubricants, pigments, antistatic agents, light stabilizers, antioxidants, and inorganic bulking agents. Examples of such other lubricants include materials based on aliphatic acids, aliphatic amides, aliphatic bis-amides, aliphatic ketones, and higher alcohols. In addition, the following materials can be used as pigments: organic dyes such as phthalocyanine blue, phthalocyanine green, isoindolinone yellow, quinacridone red, perylene red; and inorganic dyes such as ultramarine blue, cobalt blue, chromium oxide green, titanium white, carbon black, red ocher, cadmium yellow, and cadmium red. Furthermore, cationic systems, anionic systems, and non-ionic systems can be used as antistatic agents. Benzophenone systems, benzotriazole systems, hindered amine systems, and salicylic ester systems can be used as light stabilizers. Calcium carbonate, aluminum hydroxide, magnesium hydroxide, talcum, feldspar, silica, hydrotalcite, and the like can be used as inorganic bulking agents.

**[0011]** The polyester resin sheeting of the present invention is obtained by means of a method in which the copolyester resin, fatty acid ester lubricant, and optional additives are blended in appropriate amounts, kneaded together to prepare a raw material, and molded into a sheet by means of a conventional calendering method. Alternatively, the sheeting can be obtained by means of a method in which a so-called master batch containing a fatty acid ester lubricant and optional additives blended in high concentrations with the copolyester resin is blended with the copolyester resin in a specific amount to obtain a specific blending ratio, and the resulting material is kneaded and formed into sheeting by means of the same calendering method as that described above.

**[0012]** When the above-mentioned polyester resin is processed into a sheet, one or both surfaces will be embossed, but if the surface roughness  $R_a$  is less than 0.5  $\mu\text{m}$ , blocking will occur in the sheeting, and problems will be encountered during card fabrication. Conversely, if the surface roughness exceeds 15  $\mu\text{m}$ , printing-related performance attributes such as printing ink adhesion,

ink relief finish, and printer applicability will be compromised. Therefore, the surface roughness Ra is set to 0.5 to 15  $\mu\text{m}$ , and preferably 1 to 10  $\mu\text{m}$ . There are no particular limitations as to the embossing method used; embossing may be performed as a result of using the temperature conditions of the rolls during calendering, using embossing rolls wherein the surface of the final roll has been sand-matted or engraved, or passing a preheated sheeting between an embossing roll and a pressure roll.

**[0013]** If the sheeting according to the present invention is used as a card core material, a white pigment is added to the above-mentioned polyester resin composition in order to impart masking properties to the sheeting, but the masking properties will be insufficient if the added amount is less than one weight part of the white pigment per 100 weight parts of the above-mentioned copolyester resin, and if the amount exceeds 20 weight parts, processability will be compromised and the sheeting will become difficult to process. Consequently, the white pigment is used in an amount of 1 to 20 weight parts, preferably, 3 to 18 weight parts. There are no particular limitations as to the white pigment used, with the following inorganic compounds able to be used as such pigments: titanium oxide, barium sulfate, calcium carbonate, aluminum hydroxide, magnesium hydroxide, talcum, and hydrotalcite.

**[0014]**

**[Working Examples]** The present invention is described in detail below with reference to working examples; however, the present invention shall not be construed to be limited to these examples.

**(Working Examples 1 to 7)**

A copolyester resin in which the ethylene glycol component of a polyethylene terephthalate resin had been substituted with 30 mol% cyclohexanedimethanol was used. The raw material was obtained by means of blending stearic acid monoglyceride, montanic acid ester, and montanic acid ester partially saponified with calcium as fatty acid ester lubricants in the proportions shown in Table 1 (with the numbers designating weight parts). Polyester resin sheetings with a thickness of 0.3 mm were manufactured by means of conventional calendering at 160°C. The condition of roll lubricity, the transparency of the resulting polyester resin sheetings, and other appearance attributes were observed during the calendering operation. The results are shown in Table 1.



[0015]

[Table 1]

	Working examples						
	1	2	3	4	5	6	7
Copolyester resin	100	100	100	100	100	100	100
Stearic acid monoglyceride	0.2	0.8	1.8				
Montanic acid ester				0.9	1.9		
Partially Ca-saponified montanic acid ester						0.7	1.5
Roll lubricity	△	○	○	○	○	○	○
Transparency and other appearance traits	○	○	△	○	△	○	△

Roll lubricity:                      ○      no roll adhesion (the sheeting did not adhere to the roll)  
    △      some adhesion to the rolls, but no problems encountered in operation  
    ×      significant adhesion to the rolls; calendar molding impossible

Transparency and other  
appearance traits:                      ○      good transparency and the like; satisfactory appearance  
    △      some degradation of transparency or the like, but no practical problems  
    ×      transparency and the like compromised; sheetings unusable

[0016] (Comparative examples 1 to 7)

The sheeting used in the comparative examples was manufactured in the same manner as described in Working Examples 1 to 7 from a material obtained by means of blending 100 weight parts of a copolyester resin (in which the ethylene glycol component of polyethylene terephthalate resin had been substituted in the various amounts (mol%) shown in Table 2) with montanic acid ester, stearic acid, calcium soap, and polyethylene wax as lubricants in the proportions shown in Table 2 (with the numbers designating weight parts). The conditions of roll lubricity, transparency, and other appearance attributes were observed during the calendaring operation in a manner similar to the one described in Working Examples 1 to 7. The results are shown in Table 2.

[0017]

[Table 2]

	Comparative examples						
	1	2	3	4	5	6	7
Amt. of substituted ethylene glycol component (%)	30	30	30	30	30	3	60
Montanic acid ester		3				0.7	0.7
Stearic acid monoglyceride			0.7				
Calcium soap				0.7			
Polyethylene wax					0.7		
Roll lubricity	×	○	×	×	×	×	×
Transparency and other appearance traits	—	×	—	—	—	—	—

Roll lubricity:

same as in Table 1

Transparency and other appearance traits:

same as in Table 1

**[0018] (Working Examples 8 to 10 and Comparative Examples 8 to 9)**

The raw material used in working examples 2, 4, and 6 was calendered in the same manner as in the working examples, the sheetings were preheated at 150°C, and the preheated sheetings were passed between an embossing roll and a pressure roll and embossed under variable conditions, yielding card sheetings. Table 3 shows evaluation results for the surface roughness Ra of the sheetings, blocking properties, and printability.

[0019]

[Table 3]

	Working examples			Comparative examples	
	8	9	10	8	9
Copolyester resin	100	100	100	100	100
Stearic acid monoglyceride	0.8				
Montanic acid ester		0.9		0.9	0.9
Ca partially saponified montanic acid ester			0.7		
Surface roughness Ra ( $\mu$ )	6	6	8	0.4	20
Blocking properties	○	○	○	×	○
Printability	○	○	○	○	×

Blocking properties:

- no blocking between sheetings; no problems encountered
- △ some blocking occurs, but sheetings can still be used
- ×

Printability:

- no problems encountered in terms of printing ink adhesion or ink relief finish; good printability
- △ some problems are encountered in terms of ink adhesion and ink relief finish, but printing is still possible
- ×

[0020] (Working examples 11 to 13 and Comparative examples 10 to 11)

Copolyester resins in which the ethylene glycol component of the polyethylene terephthalate resin was substituted with 30 mol% cyclohexanedimethanol were used, and montanic acid ester (as a lubricant) and titanium oxide (as a white pigment) were added in the amounts shown in Table 4 (with the numbers designating weight parts). Sheetings were manufactured in the same manner as described in Working Examples 1 to 7, and core materials for card sheetings were produced. Evaluation results for the masking properties and processability of the sheetings are shown in Table 4.

[0021]

[Table 4]

	Working examples			Comparative examples	
	11	12	13	10	11
Copolyester resin	100	100	100	100	100
Montanic acid ester	0.9	0.9	0.9	0.9	0.9
Titanium oxide	2.5	10	19	0.5	25
Masking properties	△	○	○	×	○
Sheeting processability	○	○	△	○	×

- Masking properties:
- no problems encountered in terms of masking properties; sheetings usable
  - △ masking properties are somewhat inadequate, but the sheetings can still be used
  - ×
- Sheeting processability:
- no problems are encountered during calendaring; sheetings can be processed
  - △ some problems are encountered, but the sheetings can still be processed
  - ×
- there are no masking properties; sheetings unusable
- sheetings cannot be processed

[0022]

**[Effect of the Invention]** According to the present invention, the production of polyester resin sheetings by means of calendaring, which has been hitherto been impossible, can be accomplished, and large sheeting can be manufactured at a higher rate in comparison with the rate of sheeting produced using conventional extrusion. In addition, the processing temperature in calendaring can be reduced and the heat history shortened in comparison with conventional extrusion, thereby minimizing any deterioration in shock resistance and other physical properties due to the hydrolysis of the polyester resin.